

REMARKS / ARGUMENTS

Remarks Regarding Informalities and Claims Rejected Under 35 USC §112

Claims 1 – 25 remain in the application. Claims 35 – 46 have been added to the application and are presented for examination. Claims 26 – 34 have been withdrawn in response to a requirement by the Examiner that the claims be restricted to Claims 1 – 25, drawn to a catalyst, or to Claims 26 – 34, drawn to a process of preparing a catalyst. In view of the Examiner's earlier restriction requirement, applicant retains the right to present claims 26 – 34 in a divisional application.

The Examiner objected to claims 1, 11, 16, 22, and 23 because of noted informalities. As proposed by the Examiner, claims 1 and 11 have been reworded in lines 1 – 3. Further, as proposed by the Examiner: in claim 1, line 5, “being” has been replaced with “are”; in claim 11, line 2, “being” has been replaced with “is”; and, in claim 16, line 2, “molecules” has been deleted to retain consistency with the language of claim 15. Further formality amendments include: in claim 22, line 1, “being coated” has been deleted and replaced with “coating”; and in claim 23, line 2, “being” has been replaced with “is” and “wherein” has been added. The amendments of claim 22 and claim 23 were suggested by the Examiner but required further correction than proposed to have grammatically correct sentences after amendment. Finally, in claim 11, step (c), “essentially completely” has been deleted to address a concern raised by the Examiner with the original language. No new subject matter has been added in making these amendments.

The Examiner objected to claim 17 based on the failure to comply to 37 CFR 1.75(c). The applicant respectfully challenges this objection. Claim 17 depends from claim 16 which recites a cobalt (II) complex having “coordination sphere ligands selected from the group consisting of water, ammonia, ...” Claim 17 recites “The catalyst of Claim 16 wherein said cobalt (II) complex is hexaammine cobalt (II) carbonate.” As is known in the art, the “hexaammine cobalt (II) carbonate” coordination sphere ligands are ammonia, as recited in claim 16. The carbonate is not a coordination sphere ligand but rather a counterion to the positively charged cobalt ion. Thus, applicant requests the Examiner to reconsider his objection with respect to claim 17.

Claims 1 – 10 and 16 – 17 were rejected under 35 U.S.C. §112. The Examiner states that Claim 1 recites the limitation “the metal oxide” with insufficient antecedent basis for this limitation. The applicant respectfully notes that in the specification on page 4, lines 2 – 3, the catalyst has “a metal oxide crystallite size of from about 40 Å to about 200 Å.” On page 12, lines 11 – 15, the metal oxide is more narrowly defined as the “cobalt oxide crystallite size” – and as is known in the art, cobalt is considered a metal when in reference to oxides – and the “cobalt oxide crystallite size ... is greater than about 40 Å diameter, and is preferably less than about 200 Å.” Thus, applicant requests the Examiner to reconsider his objection under 35 U.S.C. §112 with respect to claim 1, and its dependent claims 2 – 10. If it would expedite matters, the applicant is willing to amend the paragraph on page 12 to include the more inclusive language of page 4 if so requested by the Examiner.

As noted by the Examiner, Claim 6 included the indefinite phrase “such as”. Claim 6 has been amended to delete this phrase and should now be in allowable form.

As noted by the Examiner, Claim 16 included improper Markush terminology. Claim 16 has been amended to be consistent with the correct Markush phrase, and claim 16 and its dependent claim 17 should now be in allowable form.

Remarks Regarding Allowable Subject Matter

The Examiner has indicated that Claim 22 is objected to as being dependent on a rejected base claim, but would be allowable if rewritten in independent form including all of the limitations of the base claim and any intervening claims. In response to this rejection, new claim 35 is presented. New claim 35 includes the limitations of rejected independent claims 11 (preamble and steps a – g), rejected dependent claim 21 and objected to dependent claim 22 (step h). Claims 36 – 46 have been added as claims dependent on new independent claim 35. As amended, independent claim 35 is believed to now be in allowable form. Because they depend from an allowable claim, dependent claims 36 – 46 are also believed to now be in allowable form.

The Examiner has also indicated that claims 1 – 10 would be allowable if the 35 U.S.C. §112 rejection is overcome. As noted in the prior section, page 4, lines 2 – 3, of the specification teach a catalyst that has “a metal oxide crystallite size of from about 40 Å to about 200 Å” and page 12, lines 11 – 15, of the specification teach “cobalt oxide crystallite size ... greater than about 40 Å diameter, and is preferably less than about 200 Å.” Applicant is willing to amend the paragraph on page 12 to include the more inclusive language of page 4 if so requested by the Examiner.

Remarks Regarding Citations

Applicant has made note of the prior art recited by the Examiner in Paragraph 15 of his Office Action mailed March 26, 2003.

Remarks Regarding Claims Rejected Under 35 USC §102(b) and 35 USC §103(a)

The Examiner has rejected independent claim 11 and dependent claims 12, 14 – 21, 23 and 25 under 35 U.S.C. 102(b) as anticipated by or, in the alternative, under 35 U.S.C. 103(a) as obvious over Shen et al., U.S. Patent 5,962,367 (“the ‘367 patent”). Applicant respectfully traverses both rejections.

The Examiner has further rejected independent claim 11 and dependent Claims 12 – 13, 15 – 21 and 23 – 25 under 35 U.S.C. 102(b) as anticipated by or in the alternative, under 35 U.S.C. 103(a) as obvious over Sapienza et al., U.S. Patent 4,396,539 (“the ‘539 patent”). Applicant respectfully traverses both rejections.

Summary of the Present Invention

The Fischer-Tropsch catalyst of the present invention is a transition metal-based catalyst having a high surface area, a smooth, homogeneous surface morphology, an essentially uniform distribution of cobalt throughout the support, and a small metal oxide crystallite size. In a first embodiment, the catalyst has a surface area of from about 100 m²/g to about 250 m²/g; an essentially smooth, homogeneous surface morphology; an essentially uniform distribution of metal throughout an essentially inert support; and a metal oxide crystallite size of from about 40 Å to about 200 Å. Optionally, the catalyst may further comprise at least one promoter.

The Fischer-Tropsch catalyst of independent claim 11 of the present application, as amended, is a catalyst comprising cobalt dispersed on a support to form a catalyst particle having a high surface area, a smooth, homogeneous surface morphology, an essentially uniform distribution of cobalt throughout the support and a small metal oxide crystallite size, and wherein the particle is formed by several prescribed steps. Claims 12 – 25 of the present application depend from independent claim 11.

As provided in dependent claim 14, the catalyst of claim 11 may be prepared from a support having a particle size of from about 60 µm to about 150 µm, a surface area of from about 90 m²/g to about 210 m²/g, a pore volume of from about 0.35 ml/g to about 0.50 ml/g, and a pore diameter of from about 8 nm to about 20 nm. However, as discussed on page 28, line 1 through page 29, line 4 and as shown in Figures 1A – 4, two catalysts prepared with the same support material can have significantly different physical characteristics depending upon the process used to make the catalyst. (N.B.: In the examples of the present application each catalyst was prepared using a Puralox SCCa 5/150 support having a surface area of about 160 m²/g, a pore volume of about 0.50 ml/g, and a pore diameter of about 12.55 nm.) The steps outlined in independent claim 11 result in a catalyst particle having the desired physical characteristics. Prior art methods of catalyst particle preparation, even starting with the same support, do not result in the catalyst particle of the present invention.

Summary of U.S. Patent 5,962,367, Shen et al.

U.S. Patent 5,962,367 teaches a process for preparing a catalyst support primarily formed of titania. The catalyst support comprises 60 wt% to 100 wt % titania as TiO₂ and 0 wt% to 40 wt % alumina as Al₂O₃. As noted by the Examiner, the ‘367 patent teaches a cobalt molybdate catalyst supported on a titania support, wherein the support has a surface area ranging from 80 m²/g to 200 m²/g, a pore volume of from about 0.3 ml/g to about 0.5 ml/g, and a pore diameter of from about 6 nm to about 20 nm.

The ‘367 patent then teaches using this catalyst support to prepare a cobalt molybdate catalyst using an impregnation process. At column 5, lines 6 – 8, the ‘367 states “the dynamic co-impregnation

method is used for supporting the cobalt and molybdenum active components ..." In Examples 4 - 7, the support is "immersed into the impregnating solution ..." for the preparation of the catalyst.

However, the '367 patent does not teach or suggest that the catalyst resulting from the impregnation process using the '367 titania support has a high surface area, a smooth, homogeneous surface morphology, an essentially uniform distribution of cobalt throughout the support and a small metal oxide crystallite size as is seen with the preparation process of the present invention. Rather, there is nothing in the '367 patent that suggests that the physical characteristics of the cobalt molybdate catalyst prepared by the impregnation process would be any different than the catalysts prepared by the prior art impregnation method and reported in Example 6 (and Figures 2A, 2B and 4) of the present application. Further, the catalyst of the present invention made by the process of instant claim 11 would not be anticipated or obvious merely because a titania support is used, or the physical characteristics of the support are similar to supports of the prior art, or because a promoter is added. Thus, independent claim 11 and its dependent claims 12 - 25 are not anticipated nor obvious in view of U.S. Patent 5,962,367.

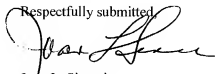
Summary of U.S. Patent 4,396,539, Sapienza et al.

U.S. Patent 4,396,539 teaches a process for preparing a catalyst suitable for Fischer-Tropsch synthesis wherein the catalyst is composed of palladium or platinum and cobalt supported on a solid support. More specifically, the '539 patent teaches an alumina, silica gel, kieselguhr, or zinc oxide support that is impregnated by immersion of the support in an aqueous solution of the salt of a palladium or platinum metal. The palladium- or platinum-treated support is then immersed in a solution containing primarily cobalt.

The objective of the process set forth in the '539 patent is to achieve a catalyst particle with the metal loading as a coating around the support. The catalyst structure depicted in Figure 5 is believed to "best represent the idealized structure of this catalyst" (column 4, lines 9 - 21). By contrast, the process of claim 11 of the present invention produces a catalyst that has "an essentially uniform distribution of cobalt throughout the support". In other words, the process taught in the '539 patent teaches away from the desired product of the present invention. Thus, independent claim 11 and its dependent claims 12 - 25 are not anticipated nor obvious in view of U.S. Patent 4,396,539.

Because U.S. Patent 5,962,367 and U.S. Patent 4,396,539 each teach a process for preparing a catalyst that relies on impregnation to deliver the metal to the support, the product-by-process taught by the amended claim 11 of the present application is neither anticipated nor obvious. Thus, applicant respectfully requests that the §102(b) and the §103(a) rejections be withdrawn and that independent claim 11, as amended, and its dependent claims 12 - 25, be allowed.

Applicant respectfully requests that a timely Notice of Allowance be issued in this case.

Respectfully submitted,

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Clean Unmarked Version of Claims Now in Application

Claim 1. A catalyst for use in the Fischer-Tropsch process, said catalyst comprising a catalyst particle, which comprises at least one metal that is an efficient carbon monoxide adsorber and at least one promoter dispersed on a support to form said catalyst particle, said particle having a BET surface area of from about 100 m²/g to about 250 m²/g, and said metal and said promoter are dispersed on the support such that the crystallite size of the metal oxide is from about 40 Å to about 200 Å, and said particle having an essentially smooth, homogeneous surface morphology.

Claim 2. The catalyst of Claim 1 wherein said particle comprises from about 5 wt % to about 60 wt % cobalt, and from about 0.0001 wt % to about 1 wt % of a first promoter, and from about 0.01 wt % to about 5 wt % of a second promoter.

Claim 3. The catalyst of Claim 2 wherein said particle comprises from about 10 wt% to about 30 wt % cobalt, and from about 0.01 wt % to about 0.05 wt % of said first promoter, and from about 0.1 wt % to about 1 wt % of said second promoter.

Claim 4. The catalyst of Claim 1 wherein said metal is selected from the group consisting of nickel, cobalt, iron, ruthenium, osmium, platinum, palladium, iridium, rhenium, molybdenum, chromium, tungsten, vanadium, rhodium, copper, zinc, and combinations thereof.

Claim 5. The catalyst of Claim 4 wherein said metal is cobalt.

Claim 6. The catalyst of Claim 1 wherein said promoter is selected from the group consisting of boron, cerium, chromium, copper, iridium, iron, lanthanum, manganese, molybdenum, palladium, platinum, rhenium, rhodium, ruthenium, strontium, tungsten, vanadium, zinc, sodium oxide, potassium oxide, rubidium oxide, cesium oxide, magnesium oxide, titanium oxide, zirconium oxide, scandium, yttrium, praseodymium, neodymium, promethium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, lutetium, other rare earth metals and combinations thereof.

Claim 7. The catalyst of Claim 2 wherein said first promoter is selected from the group consisting of palladium, platinum, ruthenium, rhenium, rhodium, iridium and a combination thereof; and said second promoter is selected from the group consisting of potassium, boron, cesium, lanthanum, cerium, strontium, scandium, yttrium, praseodymium, neodymium, promethium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, lutetium, palladium, platinum, ruthenium, rhenium, rhodium, iridium and combinations thereof.

Claim 8. The catalyst of Claim 1 wherein said support is selected from the group consisting of aluminum oxide, γ -alumina, alumina monohydrate, alumina trihydrate, alumina-silica, magnesium silicate, silica, silicate, silicalite, γ -zeolite, mordenite, titania, thoria, zirconia, niobia, hydrocalcite, kieselguhr, attapulgite clay, zinc oxide, other clays, other zeolites and combinations thereof.

Claim 9. The catalyst of Claim 8 wherein said support is γ -alumina.

Claim 10. The catalyst of Claim 9 wherein said support has a particle size of from about 60 μ m to about 150 μ m, a surface area of from about 90 m²/g to about 210 m²/g, a pore volume of from about 0.35 ml/g to about 0.50 ml/g, and a pore diameter of from about 8 nm to about 20 nm.

Claim 11. A catalyst for use in the Fischer-Tropsch process, said catalyst comprising cobalt dispersed on a support to form a catalyst particle having a high surface area, a smooth, homogeneous surface morphology, an essentially uniform distribution of cobalt throughout the support and a small metal oxide crystallite size, and wherein said particle is formed by the steps of:

- a) adding said support to water, with agitation, to form a slurry, and maintaining a slurry temperature at from about 35°C to about 210°C;
- b) adding an aqueous cobalt salt solution having a pH value greater than the point of zero charge of said support to said slurry with agitation and while maintaining said slurry temperature at from about 65°C to about 120°C;
- c) agitating said slurry and maintaining said slurry temperature at from about 65°C to about 120°C until said cobalt salt is reacted with said support;
- d) separating said slurry into a solid portion and a liquor portion;
- e) washing said solid portion with water;
- f) drying and calcining said solid portion at from about 90°C to about 375°C to form catalyst particles; and
- g) reducing said catalyst particles by heating said particles from ambient temperature to from about 300°C to about 500°C at a rate of from about 0.1°C/min to about 10°C/min over a period of from about 5 hours to about 40 hours.

Claim 12. The catalyst of Claim 11 wherein said support is selected from the group consisting of aluminum oxide, γ -alumina, alumina monohydrate, alumina trihydrate, alumina-silica, magnesium silicate, silica, silicate, silicalite, γ -zeolite, mordenite, titania, thoria, zirconia, niobia, hydrocalcite, kieselguhr, attapulgite clay, zinc oxide, other clays, other zeolites and combinations thereof.

Claim 13. The catalyst of Claim 12 wherein said support is aluminum oxide.

Claim 14. The catalyst of Claim 12 wherein said support has a particle size of from about 60 μm to about 150 μm , a surface area of from about 90 m^2/g to about 210 m^2/g , a pore volume of from about 0.35 ml/g to about 0.50 ml/g , and a pore diameter of from about 8 nm to about 20 nm .

Claim 15. The catalyst of Claim 11 wherein said cobalt salt solution comprises water and a cobalt (II) complex having coordination sphere ligands selected from the group consisting of water, chloride ion, ammonia, pyridine, triphenylphosphine, 1,2-diaminoethane, diethylenetriamine, triethylenetetraamine, acetate, oxalate, 2,4-pentanedione, ethylenedinitilo tetraacetic acid, and combinations thereof.

Claim 16. The catalyst of Claim 15 wherein said cobalt (II) complex has coordination sphere ligands selected from the group consisting of water, ammonia, pyridine, diaminoethane, diethylenetriamine, triethylenetetraamine, and a combination thereof.

Claim 17. The catalyst of Claim 16 wherein said cobalt (II) complex is hexaammine cobalt (II) carbonate.

Claim 18. The catalyst of Claim 11 wherein the slurry temperature is maintained at from about 65°C to about 120°C in step a).

Claim 19. The catalyst of Claim 11 wherein said solid portion is dried at from about 120°C to about 260°C in step f).

Claim 20. The catalyst of Claim 11 wherein said particles are reduced in step g) by heating said particles from ambient temperature to about 350°C at a rate of about 1.0°C/min and then holding said particles at about 350°C for from about 12 hours to about 16 hours.

Claim 21. The catalyst of Claim 11 wherein said particles are further stabilized to prevent pyrophoric reactions when said particles are in the presence of air.

Claim 22. The catalyst of Claim 21 wherein said particles are stabilized by coating with oil.

Claim 23. The catalyst of Claim 11 further including at least one promoter wherein said promoter is added with said cobalt salt solution.

Claim 24. The catalyst of Claim 23 wherein said promoter is a metal salt selected from the group consisting of rhenium (VII) oxide, ruthenium nitrosyl nitrate, platinum chloride, platinum ammine nitrate, platinum ammine chloride, and combinations thereof.

Claim 25. The catalyst of Claim 11 further including at least one promoter impregnated onto said catalyst particle after said particle is dried in step f), said promoter being impregnated onto said particle by dipping said particle in an aqueous solution of said promoter while maintaining agitation, and then separating said impregnated particles from said solution, and drying said impregnated particles.

Claim 35. A catalyst for use in the Fischer-Tropsch process, said catalyst comprising cobalt dispersed on a support to form a catalyst particle, wherein said particle is formed by the steps of:

- a) adding said support to water, with agitation, to form a slurry, and maintaining a slurry temperature at from about 35°C to about 210°C;
- b) adding an aqueous cobalt salt solution having a pH value greater than the point of zero charge of said support to said slurry with agitation and while maintaining said slurry temperature at from about 65°C to about 120°C;
- c) agitating said slurry and maintaining said slurry temperature at from about 65°C to about 120°C until said cobalt salt is reacted with said support;
- d) separating said slurry into a solid portion and a liquor portion;
- e) washing said solid portion with water;
- f) drying and calcining said solid portion at from about 90°C to about 375°C to form catalyst particles;
- g) reducing said catalyst particles by heating said particles from ambient temperature to from about 300°C to about 500°C at a rate of from about 0.1°C/min to about 10°C/min over a period of from about 5 hours to about 40 hours; and
- h) stabilizing said particles to prevent pyrophoric reactions when said particles are in the presence of air by coating said particles with oil.

Claim 36. The catalyst of Claim 35 wherein said support is selected from the group consisting of aluminum oxide, γ -alumina, alumina monohydrate, alumina trihydrate, alumina-silica, magnesium silicate, silica, silicate, silicalite, γ -zeolite, mordenite, titania, thoria, zirconia, niobia, hydrotalcite, kieselguhr, attapulgite clay, zinc oxide, other clays, other zeolites and combinations thereof.

Claim 37. The catalyst of Claim 36 wherein said support has a particle size of from about 60 μm to about 150 μm , a surface area of from about 90 m^2/g to about 210 m^2/g , a pore volume of from about 0.35 ml/g to about 0.50 ml/g , and a pore diameter of from about 8 nm to about 20 nm.

Claim 38. The catalyst of Claim 35 wherein said cobalt salt solution comprises water and a cobalt (II) complex having coordination sphere ligands selected from the group consisting of water, chloride ion, ammonia, pyridine, triphenylphosphine, 1,2-diaminoethane, diethylenetriamine, triethylenetetraamine, acetate, oxalate, 2,4-pentanedione, ethylenedinitilo tetraacetic acid, and combinations thereof.

Claim 39. The catalyst of Claim 38 wherein said cobalt (II) complex has coordination sphere ligands selected from the group consisting of water, ammonia, pyridine, diaminoethane, diethylenetriamine, triethylenetetraamine, and a combination thereof.

Claim 40. The catalyst of Claim 35 wherein the slurry temperature is maintained at from about 65°C to about 120°C in step a).

Claim 41. The catalyst of Claim 35 wherein said solid portion is dried at from about 120°C to about 260°C in step f).

Claim 42. The catalyst of Claim 35 wherein said particles are reduced in step g) by heating said particles from ambient temperature to about 350°C at a rate of about 1.0°C/min and then holding said particles at about 350°C for from about 12 hours to about 16 hours.

Claim 43. The catalyst of Claim 35 further including at least one promoter wherein said promoter is added with said cobalt salt solution.

Claim 44. The catalyst of Claim 43 wherein said promoter is a metal salt selected from the group consisting of rhenium (VII) oxide, ruthenium nitrosyl nitrate, platinum chloride, platinum ammine nitrate, platinum ammine chloride, and combinations thereof.

Claim 45. The catalyst of Claim 35 further including at least one promoter impregnated onto said catalyst particle after said particle is dried in step f), said promoter being impregnated onto said particle by dipping said particle in an aqueous solution of said promoter while maintaining agitation, and then separating said impregnated particles from said solution, and drying said impregnated particles.

Claim 46. The catalyst of Claim 35 wherein said support is aluminum oxide and said aqueous cobalt salt solution comprises water and hexaammine cobalt (II) carbonate.